



Moth bean starch (*Vigna aconitifolia*): isolation, characterization, and development of edible/biodegradable films

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Abstract In the current work, moth bean starch was separated from the moth bean seeds which were cultivated in the semi arid regions of Haryana and Rajasthan, India using alkali treatment technique and characterized. Isolated and purified moth bean starch comprised (12.89–20.37%) amylose, 0.8% protein, 0.4% ash, swelling index and solubility were 10.8–14.7% and 6.4–9.8% respectively. For wrapping application, starch was modified using citric acid as cross linking agent (1–7% w/w of total starch) and film was made using casting method, and investigated the influence of citric acid on the functional properties of the films such as moisture content, solubility, swelling index, water vapor permeability and mechanical property. Significant difference in the functional properties among native and modified moth bean starch films was noticed. Interactions among starch chains due to cross linking in the modified starch films were reported using FTIR spectra. Surface micrographs of both purified starch granules and films were studied using scanning electron microscopy. From the outcomes, it was exhibited that obtained starch granules has range large to small size with distorted cylinder and oval shapes. While cross linked starch films showed compact, slightly rough and homogeneous surface. Tested modified moth bean starch films containing citric acid (CA) and sorbitol were utilized as wrapping films to improve the shelf life of fresh lemon. Outcomes showed

that the film contains 5% CA could be most promising wrapping materials for lemon that enhanced the shelf life of lemon additionally up to 12 days.

Keywords Moth bean starch · Citric acid · Barrier properties · Shelf life · Lemon quality

Introduction

Currently, the polymers obtained from the renewable sources such as agricultural waste, marine waste, and food industry waste have been utilized as a new strategy to decrease the severe environmental problems resulted by consumption of synthetic packaging materials. Edible films are generally prepared from renewable natural polymers such as polysaccharides, resin, proteins, lipids or waxes or mixtures thereof (Ferreira et al. 2018). Starch exhibited extremely well among all natural polymers for the development of biodegradable and edible films because of its ubiquitous, low cost, renewable nature and potential to form film/continuous polymeric materials. Past two decades, the population and urbanization in the developing countries has been increased. Therefore consumption of different varieties of starch has been exponentially enhanced for different applications. So, it becomes necessary to search alternative natural and renewable resources due to the shortage of the common sources of starch. In this sense, moth bean starch may be believed as one of abandon source of starch which is recently unheeded and usually utilized in Indian dishes. Moth bean starch may be assumed as most concerning source of starch which is not fully explored. It assumed to be the most cultivated crop during summer season in India. Due to low input cost and less requirement of water, this crop is cultivated in arid and

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semi arid regions in India such as Rajasthan, Haryana, Madhya Pradesh and some region of Punjab and Uttar Pradesh. It is also cultivated in other countries such as Sri Lanka, China, Africa and USA. Moth beans starch contained good amount of amylose (8–21%) (Singh et al. 2017), which is necessary for the development of biodegradable/edible films with good film properties. Instead of these properties, number of research works about development of edible or biodegradable films using moth bean starch is scant. So it becomes necessary to carry out new informative works related to its characterization of isolated starch, functional and microstructural properties of starch film, focusing to the information that contributes to its potential applications in different areas. Like, other natural polymers, starch show the feature of hydrophilicity, which support biodegradable nature. Hydrophilic character of starch also shows adverse effect on main functional properties of the films such as moisture barrier and mechanical property. To solve these practical problems, generally, various techniques were suggested such as chemical, physical and enzymatic modification methods to dilute these hindrances. Within each category of modification, considerable differences in the films properties were noticed. Chemical modifications possibly include the chemical reactions such as esterification, which are more frequently utilized than other modification approaches (Oleyaei et al. 2016). Chemical modification technique is utilized for many applications (gelling and encapsulation) to alter the micro and macroscopic features of starch matrix (Ashok kumar 2015). Starch modifications using cross linking agents such as citric acid, boric acid and oxidized sucrose is of great interest when films acquires good film strength, low moisture barrier and hydrophobic features. When starch matrix attained hydrophobic characteristic using mentioned techniques, it can be utilized in many situations, such as food packaging and pharmaceutical excipients, and others (Xu et al. 2015). Moreover, among all cross linking agents, several cross-linking agents are not consumed in food packaging because of their toxic nature. Within all cross-linking agents, most appropriate cross-linker is citric acid (CA) due to its non-toxicity, and great capabilities to enhance the film properties (Menzel et al. 2013). In this context, the purpose of this work was to develop food packaging/wrapping films by esterification reaction among the citric acid and starch chains. Influence of citric acid on the starch film functional properties such as thickness, moisture content, swelling index, solubility, opacity, mechanical and moisture barrier properties were studied to assess their potential application as wrapping films.

Materials

Moth beans were procured from local market of Chandigarh, India. Sodium hydroxide; citric acid and calcium chloride were purchased from Loba Chemical (Mumbai, India). Sorbitol was obtained from HiMedia (Mumbai, India). Fresh lemon (*Citrus limon* L.) was collected from the local vegetables markets, Chandigarh, India.

Methods

Starch isolation

Starch was isolated from moth beans using wet milling technique as reported in literature (Gunaratne et al. 2018) with minor modifications. Moth beans (100 g) were soaked in 300 mL sodium hydroxide solution (0.25% w/v of solution) at room temperature for 24 h. Thereafter, blending and dehulling of soaked moth beans was done with alkali solution using blender (Sujata, India) for 5 min. Resultant moth beans slurry was screened using muslin cloth and then followed by 200 mesh sieves. Washing of starch was recurred many times until moth bean starch seemed to be free from undesired materials. Finally, isolated starch was dried at 40 ± 1 °C for 24 h.

Film fabrication

Modified moth bean starch films were fabricated by utilizing the solvent casting method following the reported method (Kumar et al. 2019). Exactly 3% w/v of moth bean starch and sorbitol (25% w/w of total starch) were dissolved with continuous stirring using magnetic stirrer in 100 ml distilled water. Resultant starch solution was heated with continuous stirring using magnetic stirrer at 90 ± 1 °C for 10 min. Food grade citric acid (1%, 3%, 5% and 7% w/w of total dry weight of moth bean starch) was added and continued with constant heating at same conditions for another 20 min. Obtained solution was homogenized using high shear homogenizer. Resultant homogenized solutions were degassed by vacuum oven (NSW, India) and cast evenly into the petri dishes. The films were allowed to dry at 25 ± 1 °C for 24 h. Dried films were stored in air tight bag for analysis.

Characterization of starch

Amylose content of isolated starch was calculated using iodine colorimetric technique. Microstructure and crystalline pattern of starch matrix were measured using SEM (EVO15 Germany) with an acceleration potential of 15 kV

and XRD, (X'Pert PRO, Panalytical, Netherlands) with Cu K radiation at a voltage of 45 kV and 40 mA respectively. Samples were scanned in the range from 3° to 60° 2 θ with scanning speed of 2°/min. Water absorption property, swelling index, aqueous solubility and pasting property of the isolated starch were determined using previously reported methods (Lutfi et al. 2017) with small modifications.

Film characterization

Thickness

Films thickness of developed starch films was measured by employing Mitutoyo micrometer (Mitutoyo, Japan) at six different locations and then took the average for further analysis.

Moisture content

Moisture content of the films was measured according to gravimetric technique. Moisture content in terms of weight loss was evaluated using Eq. (1)

$$\text{Moisture content (MC\%)} = \frac{m_0 - m_1}{m_0} * 100 \quad (1)$$

where m_0 and m_1 are initial and dried weight of film samples respectively.

Water solubility

Solubility of the films was evaluated in terms of percentage of undissolved mass of samples in distilled water. In brief, prepared samples dried at 105 \pm 1 °C for 24 h and weighed (w_i). Dried samples were soaked in 15 ml distilled water at 25 \pm 1 °C for 24 h. Undissolved samples were removed and again dried at same conditions and weighed again (w_o). Solubility (S%) was evaluated according to the Eq. (2).

$$S(\%) = \frac{w_i - w_o}{w_i} * 100 \quad (2)$$

where w_i and w_o are initial and dried weight of insoluble samples respectively.

Swelling index

Water molecules interaction with starch polymer chains was evaluated in terms of swelling index using standard method as described in the literature with some changes (Galus and Kadzi 2016).

Light permeability

Light permeability was measured using calibrated Hunter Colorimeter (Color Flex EZ, Hunter Lab, and USA). Light permeability was evaluated as follows

$$\text{Light Permeability (LP)} = LP_b / LP_w * 100 \quad (3)$$

where LP_b and LP_w are light permeability at black and white background respectively.

Water vapor permeability

The water vapor permeability was measured using perviously reported work (Daza et al. 2018) with minor changes. In brief, samples were cut from the films and fixed on mouth of the modified glass beaker filled with dried calcium chloride with vacuum grease. All beakers were placed in the desiccators having distilled water. Desiccator then placed in controlled chamber at 25 \pm 1 °C. Increment in weight of all beakers was measured using an analytical balance (\pm 0.0001 g) over an interval of 24 h. Water vapor permeability was demonstrated using Eq. (4)

$$WVP = WVTR * Y / (p_0 - p_1) \quad (4)$$

where WVTR is water vapor migration rate through the samples, evaluated from graphs of weight gain of beakers with time, Y is the average thickness of samples and ($p_0 - p_1$) is the pressure difference between the outside and inside of beakers. Three replicates were carried out to analyze WVP for each sample.

Mechanical property

Films strength and extensibility were measured by using Texture Analyzer (TA.XT Plus, Stable Microsystems, Sury, UK) according to the procedure reported by Ghadetaj et al. (2018) with small changes.

FT-IR analysis

Interactions among the starch chains due to cross linking were visualized using FTIR (Shimadzu -8400, Japan) in the wave length range 500–4000 cm^{-1} with resolution of 4 cm^{-1} .

Microstructure analysis by scanning electron microscope (SEM)

Surface microstructure of cross linked and uncross linked moth bean starch films was carried out using a scanning electron microscope (SEM) (EVO15, Germany) with an accelerating potential of 25 kV (Ghoshal et al. 2017).

Assessment of the isolated starch films as wrapping material

Newly prepared modified starch films utilized as wrapping films on the fresh lemon and analyzed the shelf life. Lemons with similar shape, size, and colour were selected for samples. Prepared films were tightly wrapped around each sample and taped. Similarly, plastic film wrapped lemon was used as control samples. Both, plastic and modified starch films wrapped samples were then kept at control condition and samples were regularly monitored up to 12 days.

Statistical analysis

Thickness, moisture content, swelling index, opacity, water vapor permeability and mechanical properties were measured in triplicate. Results were represented as mean \pm SD and analysis was done using the Origin 8 software. Results were statistically assessed using analysis of variance (ANOVA) and multiple ranges Duncan's to determine whether effect of CA on film properties were significant at $P < 0.05$ using statistical software SPSS (version 23, SPSS Inc., Chicago, IL, USA).

Results and discussion

Characterization of isolated starch

Amylose content in starch influences the various important properties such as film forming capacity, crystallinity and hydrophilicity. In present work, amylose content in isolated starch varied widely from 10.8 to $20.9 \pm 0.41\%$ (Table 1). A comparative work illustrated that amylose content was lower-similar than that of field pea (33.2–33.6) and yam starch (11.6–23.6%), which may be due to their differences in sources, environmental and soil conditions (Zhu 2015).

Table 1 Main important properties of isolated moth bean starch properties

Sr. No.	Moth starch properties	Results
1	Amylose content (%)	20.9 ± 0.41
2	Aqueous solubility (%)	8.87 ± 0.33
3	Swelling index (g/g)	12.51 ± 0.35
4	Moisture absorption capacity (%)	75.23 ± 0.36
5	Pasting property (°C)	78 ± 0.65
6	Moisture content (%)	8.23 ± 0.25
7	Proteins content (%)	0.8 ± 0.34
8	Ash content (%)	0.4 ± 0.54

Spectrum peaks at 14.3° , 17.2° and 22.5° indicated that moth starch belongs to C type patterns. Variation in the crystalline pattern depends on many factors such as genotypes, sources and environment (Fig. 1a) which is matched with perviously published results (Ambigaipalan et al. 2011). Morphology of starch granules were carried out using SEM as shown in Fig. 1b. Starch granules were observed in small and large size with distorted cylindrical, oval and elliptical shapes. Swelling index of isolated moth starch was (12.51 g/g) at $90 \pm 1^\circ \text{C}$ which was observed to be higher than several other starch sources such as maize ($8.3\text{--}10.1 \text{ g/g}$) and wheat ($8.3\text{--}10.1 \text{ g/g}$) (Kaur et al. 2016). Likewise, moth bean starch solubility was obtained to be $8.87 \pm 0.33 \text{ g/g}$ at $90 \pm 1^\circ \text{C}$ which was also higher than rice starch. The variation in the solubility and swelling power can be assigned to the deviation in arrangement of granules in the starch and amylose content (Wani et al. 2016). Pasting temperature affected many other properties such as swelling power and solubility and was recorded to be 78°C . In our work, obtained pasting temperature of moth bean starch was higher than previously reported work on potato starch (69.7°C) (Kaur et al. 2015). Variation in the pasting temperature of starch was depended on the many factors such as amylose leaching during the heating, crystallinity, swelling power, shape and size of granules. Isolated starch contains proteins (0.8%), moisture (8.23%) and ash (0.4%) which are comparable and slightly lower

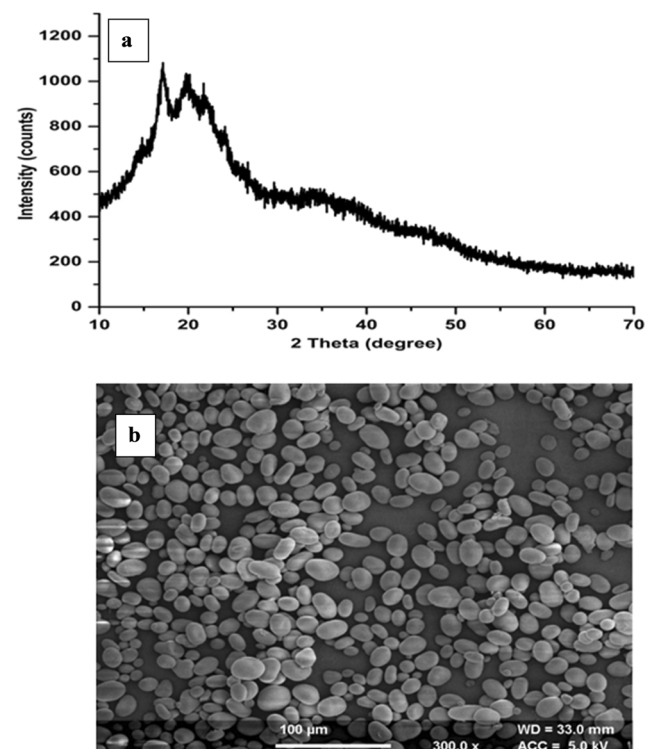


Fig. 1 a X-ray diffraction and b Scanning electron micrographs of isolated moth bean starch

than reported for other starch (Lin et al. 2012). It seems that obtained moth bean starch was pure without any impurities.

Thickness

Main important properties such as mechanical and moisture permeability of the films are the function of the film thickness. The impact of citric acid on the thickness of the films was not significant as presented in Fig. 2a. Significant difference ($P < 0.05$) in the film thickness with and without citric acid was noticed. However, addition of different amount of citric acid (1–7% w/w of total starch) caused to slightly increase in the film thickness due to increase in the degree of arrangement/orientation in the starch matrix, which slightly disturb the arrangement of starch chains in the films, causing to arise the film thickness (Azeredo and Waldron 2016). The findings were in line with previously reported work (Cuadro et al. 2015).

Moisture content (MC)

Moisture content of all prepared films is plotted in Fig. 2a. With increase of citric acid content in the films, the

moisture content of the films significantly ($P < 0.05$) reduced. It was found that when CA content (w/w of total starch) was increased from 1 to 5%, MC of the films decreased from 12.87 ± 1.2 to $8.98 \pm 0.2\%$. Similarly, Sharma et al. (2017) also observed that modified protein films illustrated lower moisture content compared to native protein films. Findings presented that cross linking strengthen the films matrix, resulted to form compact network. Besides, this can also reduce the availability of free –HO groups in the starch network. We assumed that consequently reduced moisture content results. Further incorporation of citric acid more than 5% (w/w of total starch) in the film it can be able to increase the free space in the film matrix. Therefore, mobility among the starch chains in starch matrix increased, leading to increase the MC of the films (Wang et al. 2014).

Water solubility

Water solubility of cross linked moth starch film with different concentrations of citric acid is depicted in Fig. 2b. As citric acid content in the starch films increased, water solubility significantly ($P < 0.05$) reduced. The films with

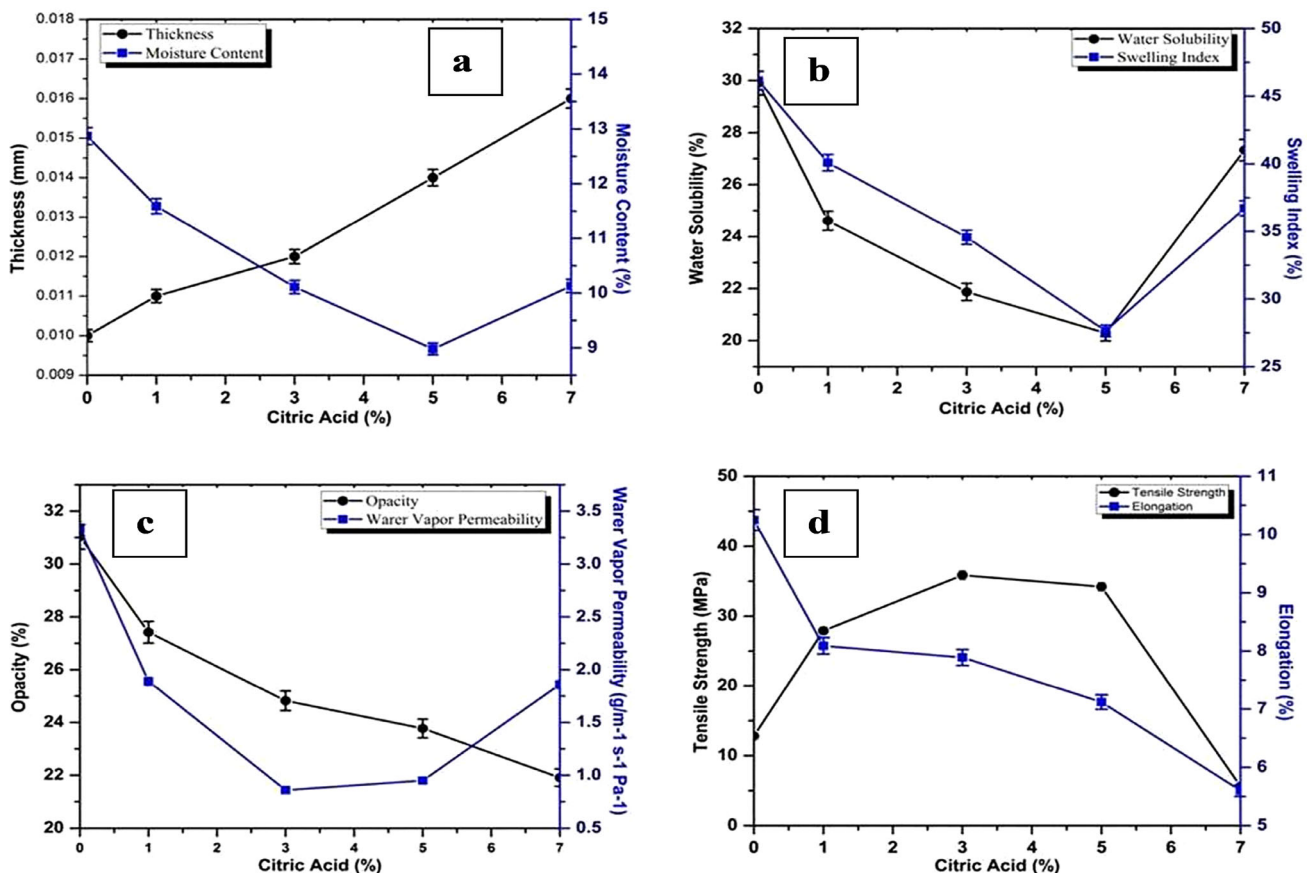


Fig. 2 Effect of citric acid on the functional properties of films **a** thickness, moisture content, **b** water solubility and swelling index, **c** opacity, water vapor permeability, and **d** tensile strength and elongation

5% citric acid (w/w of total starch) presented minimum water solubility ($20.29 \pm 0.2\%$) than other films. Diminution in the water solubility of the films can be expressed with strong fact that different amount of citric acid in the films induced cross-linking between the starch chains. Thereby, these interactions in terms of cross linking in starch chains may be reinforced in starch matrix. Consequently, compact and interlocked structure of the modified starch film was obtained, which reduced interactions towards the water of the films, leading to fell in solubility (Menzel et al. 2013). Besides these can also diminish the availability of free hydroxyl groups in the starch films caused to decrease the solubility. However, surplus CA in the films enhanced aqueous solubility of the film due to its plasticizing effect.

Swelling index

Behavior of the modified moth bean starch and unmodified moth bean starch films with water was recorded in terms of swelling index (SI) as shown in Fig. 2b. Unmodified moth bean starch film exhibited higher swelling index than modified starch film because of their hydrophilic nature. However, addition of different amount of citric acid (1–5% w/w of total starch) in the starch films caused to reduction in the swelling index. From the outcomes, it was observed that citric acid in the films showed remarkable ($P < 0.05$) effect on SI. Due to esterification reaction among the citric acid and starch in the film, some hydroxyl groups of starch matrix were replaced with hydrophobic carbonyl bond, leading to diminish hydrophilic nature of starch, consequently SI decreased. Furthermore, it notices that the modified films with more than 5% citric acid (w/w of total starch) presented higher SI than other films. The findings suggest that unanticipated citric acid in the modified starch films acted as plasticizer, causing in more hydrophilic sites in the films, consequently swelling index enhanced (Wang et al. 2014). As expected, the modified starch films with 5% citric acid showed lowest swelling index than other cross linked starch films. Cross-linking strengthened interactions among the starch chains and reduced the incursion of water molecules without distortion, similar to those suggested by Thessrimuang and Prachayawarakorn (2018).

Opacity

Packaging/coating materials prohibited light transmission through the materials and enhanced the shelf life of food products. Opacity also affect the acceptibility of fruits/vegetables. The variation in the opacity of the films with addition of citric acid content is shown in Fig. 2c. As citric acid content increased in the films, the transparency of film was significantly ($P < 0.05$) increased. While opacity of

the films decreased. Reduction in opacity of the films might be explained in terms of cross-linking among the starch molecules. These can reduce the crystallinity of starch matrix, leading to decrease the opacity. Reduction in the crystallinity of starch matrix due to cross linking was also confirmed by XRD outcomes. The findings are consistent with previous report (Saliu et al. 2018). Authors showed that transparency improved with addition of citric acid in the films.

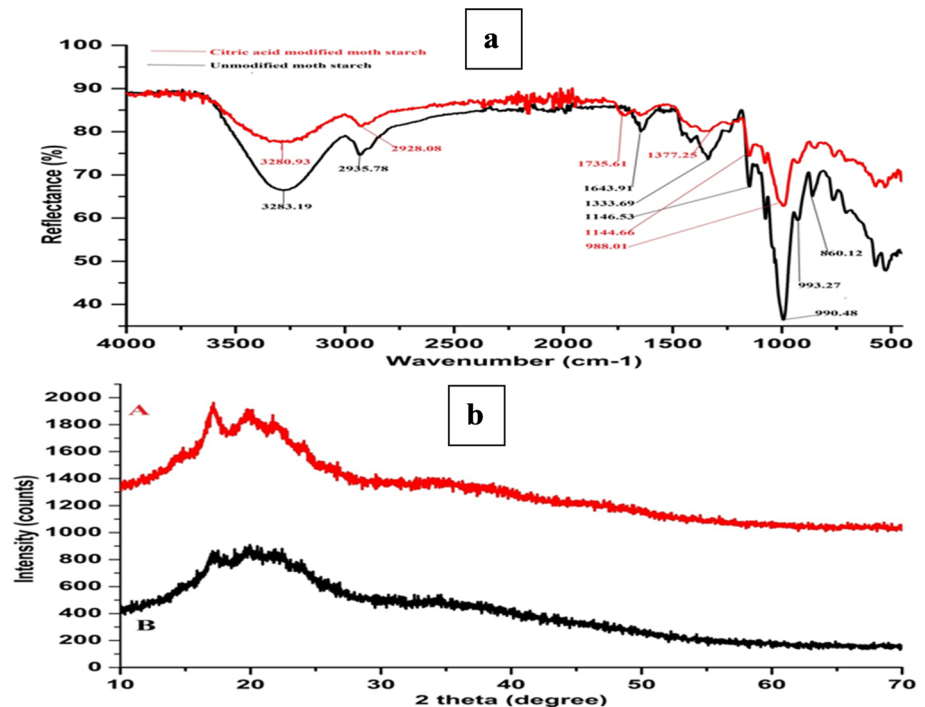
Water vapor permeability

Coating/packaging with high moisture migration rate may be created some critical problems such as food quality, shelf life and textural integrity. Therefore, moisture permeability of food packaging films should be as low as possible. The effect of varying dose of citric acid on the moisture barrier of the isolated starch film is illustrated in Fig. 2c. As citric acid dose in the starch films raised from 1 to 5% (w/w of total starch), the moisture permeability of the starch films reduced from $3.33 \pm 1.5 \times 10^{-12}$ to $0.95 \pm 1.7 \times 10^{-12} \text{ g/m}^{-1} \text{ s}^{-1} \text{ Pa}^{-1}$. The higher moisture barrier of the modified starch film might be because of low availability of free hydroxyl groups and tortuous path in the modified starch films. Mei et al. (2013) suggested that low availability of free hydroxyl groups in the starch matrix decreased interactions with water molecules/vapors, which in turn can be guided to the diminution in moisture migration rate through the films. Furthermore, addition of citric acid more than 5% in the starch films could trimmed intramolecular interactions in terms of hydrogen bonding and enhanced free space among the starch chains, which could form less dense starch network and promoted water molecules/vapor absorption in the film (Azeredo et al. 2015). The reduction in the moisture migration rate through the modified films was also because of their decreased crystallinity (Fig. 3b), which effectively decreased the density of the modified starch films and restricted to movement of water molecules/vapor through the starch films.

Mechanical property

During the transportation of food products and fruits/vegetables, packaging/coatings support them to protect from physical damages and contamination. Therefore, the film with high tensile strength can easily maintain the products integrity. Tensile strength of all the films, as illustrtaed in Fig. 2d enhanced with raising citric acid amount. As citric acid content raised from 1% to 5% (w/w of total starch), the tensile strength of the films greatly ($P < 0.05$) enhanced from 12.8 ± 0.9 to $35.86 \pm 0.8 \text{ MPa}$. Improvement in the tensile strength of the isolated moth bean starch

Fig. 3 **a** FTIR and **b** XRD analysis of citric acid modified moth starch and unmodified moth film



films could be linked with cross linking, which effectively improved the density of the films (Azeredo et al. 2016). As the citric acid content further rose from 5 to 7% (w/w of total starch), the tensile strength decreased from 35.86 ± 0.8 to 26.55 ± 1.3 MPa, which assign to plasticizing effect of citric acid. Reduction in the film strength may be evidenced that citric acid in films effectively reduced interactions among the starch chains. Consequently, movement between the starch molecules increased, leading to decrease the film strength (Menzel et al. 2013), while elongation at break point increased.

FTIR analysis

The FT-IR spectrum was utilized to identify the changes in the starch structure on a molecular level such as starch chain conformation and retrogradation when citric acid was added in the starch film. FTIR spectra of the moth bean starch and citric acid modified moth bean starch was illustrated in Fig. 3a. Both moth bean starch and modified moth bean starch demonstrated a broad peak at 3280 cm^{-1} , 3283 cm^{-1} , 2935 cm^{-1} and 2928 cm^{-1} respectively which assigned to O–H and C–H bonds stretching, respectively. The peaks around 1377 cm^{-1} and 1333 cm^{-1} could be associated with the bending structure of O–C–H, C–C–H, and C–O–H (Mei et al. 2015). Absorbance bands in the range around $1018\text{--}1155 \text{ cm}^{-1}$ were detected which related to peaks of the pyranose form of glucose residues (Ghosh et al. 2012). Absorbance peaks in the range around $850\text{--}1000 \text{ cm}^{-1}$ were ascribed in C–H bending in the modified and unmodified

moth bean starch. Additional peak in modified films at around 1735 cm^{-1} was noticed which can be assigned to the characteristic ester group (Buchanane et al. 2008). This confirms the development of ester groups due to esterification reaction among functional groups of starch (–OH) and citric acid (–COOH). A similar observation has been noted by Ma et al. (2018) who confirmed chemical modification in terms of cross-linking with the incorporation of citric acid in the soy residue. Similarly, Pornsuksomboon et al. (2016) also observed similar observation about cross-linking in the film with addition of citric acid.

XRD analysis

X-ray diffraction pattern has been widely employed to show any alteration in crystalline network of starch after chemical modification (Zobel et al. 1988). X-ray diffraction patterns of the moth bean starch and modified moth bean starch films were illustrated in Fig. 3b. Moth bean starch films established B type crystalline pattern with 2θ diffraction peak intensities at 17° , 19° and 22° (Punia et al. 2017). When citric acid was added in the moth bean starch, crystalline pattern of the modified moth bean starch film was rarely varied in comparison to the unmodified moth bean starch film except variation in the peak intensity. Intensities of these Peaks diminished after esterification reaction in the starch network occurred. This showed that cross linking exhibited in both regions (amorphous and crystalline) of starch network. These outcomes are matched with earlier published work (Zhou et al. 2016).

Scanning electron microscopic (SEM) analysis of film

This mechanism is very helpful to know the film properties. The influence of citric acid on the microstructure of the films after cross linking was investigated by SEM images as shown in Fig. 4a–d. Native starch film micrographs showed a partially smooth surface, less dense and without pore (Balakrishnan et al. 2017). While, homogeneous and compact network without any pore or crack was noticed in starch films treated with citric acid as compared with the unmodified starch films (Saliu et al. 2018). It was also observed that citric acid modified films showed more dense and homogeneous network than native starch film (Jiang et al. 2016). Similar work has been reported that cross-linked structure of whey protein depicted denser network as compared to native films.

Self life analysis of fresh lemon (food preservation application)

In order to measure the potential utilization of the prepared modified moth bean starch films in food preservation as wrapping material, lemon was wrapped with it and stored at 25 ± 1 °C and images of the samples were taken regularly during storage and were displayed in Fig. 5. At the end of experiment, it was noticed that lemon wrapped with modified moth bean starch films were still fresh and the surface remained smooth, green and without any color

variation. The weight loss of lemon wrapped with prepared films was 3%. While, control samples were slightly altered from deep green to yellowish green. From the outcomes, it was noticed that wrapping films additionally increased the shelf life of lemon up to 12 days without any loss of qualities additionally. The finding demonstrated that the developed modified starch films may be successfully used as wrapping materials which may protect food from physical damages and therefore enhance its shelf life.

Conclusion

In conclusion, starch from moth bean was successfully isolated using alkali treatment technique. The main important properties and microstructural characterization of moth bean starch were investigated. SEM outcomes of isolated moth bean starch revealed that starch granules were distorted cylindrical, oval and elliptical shapes. It was also exhibited low protein and ash content. Isolated starch showed good amylose content which essentially designate for several applications. Edible films from modified starch and unmodified starch were successfully developed using solvent casting technique. We noticed that an increase in the citric acid content (< 5% w/w of total starch content.) in the films leads to improve the functional properties such as moisture content, solubility, opacity, water vapor permeability, and swelling index. The X-ray diffraction of both modified and unmodified moth bean starch showed

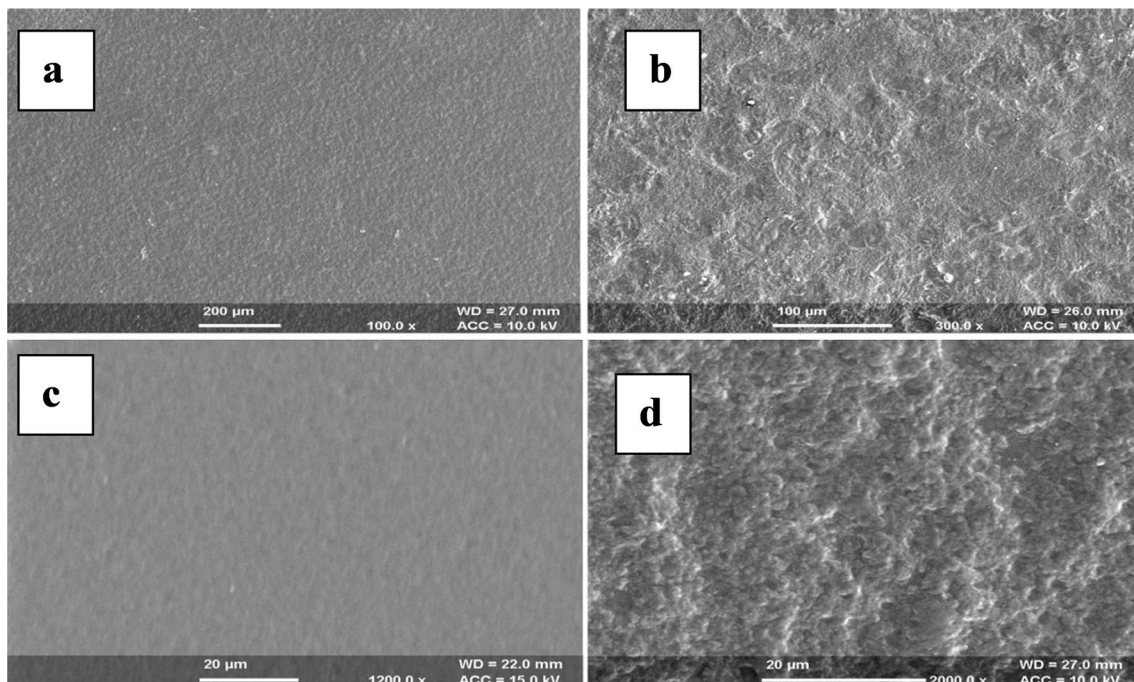
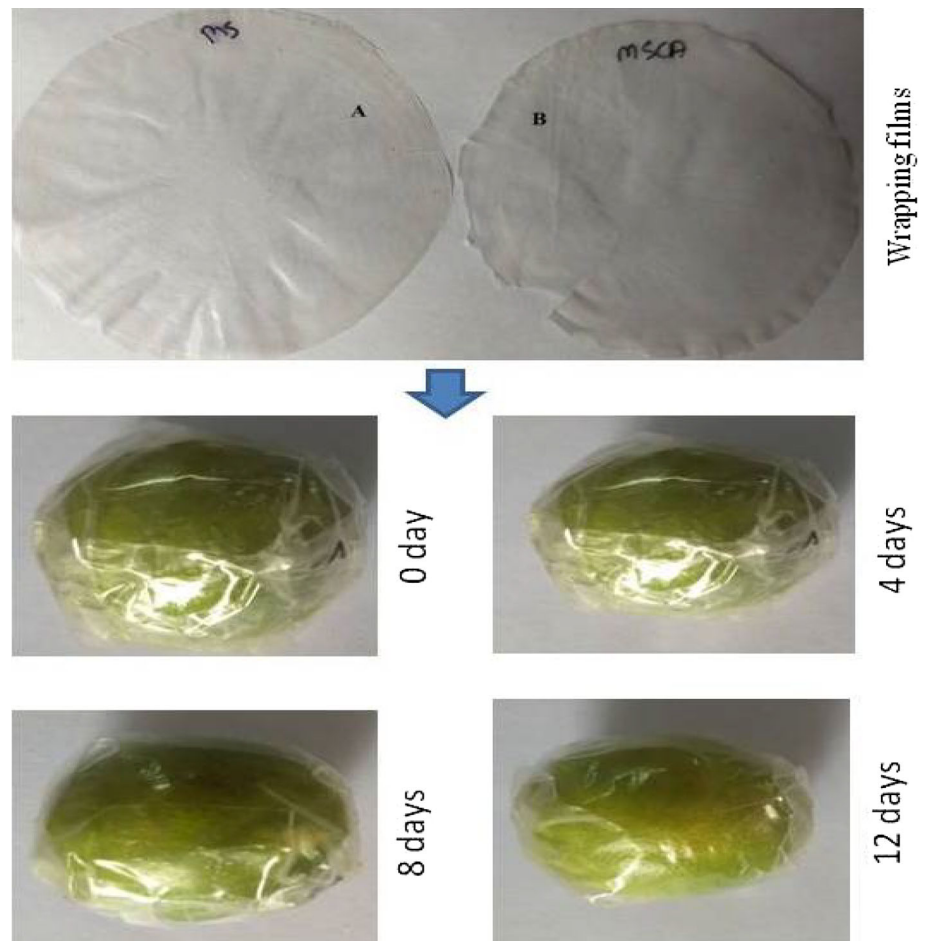


Fig. 4 SEM micrographs of (a) and (b) moth starch and (c) and (d) modified moth starch film

Fig. 5 Images of lemon covered with developed packaging materials after 4, 8 and 12 days storage at $25 \pm 1^\circ\text{C}$



that crystallinity of starch matrix diminished with incorporation of citric acid. SEM outcomes confirmed that modified films are more compact and homogeneous as compared to unmodified starch films. Modified moth bean starch films with promising functional properties enhanced the shelf life of fresh lemon up to 12 days. Moth bean starch is a very vivid natural source of starch which can be utilized for various purposes in the food applications.

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Compliance with ethical standards

Conflict of interest Authors declare no conflict of interest.

References

- Ambigaipalan P, Hoover R, Donner E et al (2011) Structure of faba bean, black bean and pinto bean starches at different levels of granule organization and their physicochemical properties. *Food Res Int* 44:2962–2974. <https://doi.org/10.1016/j.foodres.2011.07.006>
- Ashokkumar M (2015) Applications of ultrasound in food and bioprocessing. *Ultrason Sonochem* 25:17–23. <https://doi.org/10.1016/j.ultsonch.2014.08.012>
- Azeredo HMC, Waldron KW (2016) Crosslinking in polysaccharide and protein films and coatings for food contact—a review. *Trends Food Sci Technol* 52:109–122
- Azeredo HMC, Kontou-Vrettou C, Moates GK et al (2015) Wheat straw hemicellulose films as affected by citric acid. *Food Hydrocoll* 50:1–6. <https://doi.org/10.1016/j.foodhyd.2015.04.005>
- Azeredo HMC, Morrugares-Carmona R, Wellner N et al (2016) Development of pectin films with pomegranate juice and citric acid. *Food Chem* 198:101–106. <https://doi.org/10.1016/j.foodchem.2015.10.117>
- Balakrishnan P, Sreekala MS, Thomas S (2017) Morphology, transport characteristics and viscoelastic polymer chain confinement in nanocomposites based on thermoplastic potato starch and cellulose nanofibers from pineapple leaf. *Carbohydr Polym* 169:176–188
- Buchanan CM (2008) Rapid and environmentally friendly preparation of starch esters. *Carbohydr Polym* 74:137–141. <https://doi.org/10.1016/j.carbpol.2008.01.013>
- Daza LD, Homez-jara A, Solanilla JF, Vázquez HA (2018) Effects of temperature, starch concentration, and plasticizer concentration on the physical properties of ulluco (*Ullucus tuberosus* Caldas)-based edible films. *Int J Biol Macromol* 120:1834–1845. <https://doi.org/10.1016/j.ijbiomac.2018.09.211>

- De Cuadro P, Belt T, Kontturi KS et al (2015) Reactive and functional polymers cross-linking of cellulose and poly (ethylene glycol) with citric acid. *React Funct Polym J* 90:21–24. <https://doi.org/10.1016/j.reactfunctpolym.2015.03.007>
- Ferreira G, Matta F, Augustus R, Oliveira D (2018) Extraction and characterization of arrowroot (*Maranta arundinaceae* L.) starch and its application in edible films. *Carbohydr Polym* 186:64–72
- Galus S, Kadzi J (2016) Moisture sensitivity, optical, mechanical and structural properties of whey protein-based edible films incorporated with rapeseed oil. *Food Technol Biotechnol* 54:78–89
- Ghadetaj A, Almasi H, Mehryar L (2018) Development and characterization of whey protein isolate active films containing nanoemulsions of Grammosciadium pterocarpum Bioss. essential oil. *J Food Pack Shelf* 16:31–40
- Ghosh T, Netravali AN (2012) ‘Green’ crosslinking of native starches with malonic acid and their properties. *Carbohydr Polym* 90:1620–1628
- Ghoshal G, Shivhare US, Banerjee UC (2017) Rheological properties and microstructure of xylanase containing whole wheat bread dough. *J Food Sci Technol* 54(7):1928–1937
- Gunaratne A, Gan R, Wu K, Kong X, Collado L, Arachchi LV, Kumara K, Pathirana SM, Corke H (2018) Physicochemical properties of mung bean starches isolated from four varieties grown in Sri Lanka. *Starch-Stärke* 70(3–4):1700129
- Jiang S, Zhang X, Ma Y et al (2016) Characterization of whey protein-carboxymethylated chitosan composite films with and without transglutaminase treatment. *Carbohydr Polym* 153:153–159
- Kaur A, Shevkani K, Singh N, Sharma P (2015) Effect of guar gum and xanthan gum on pasting and noodle-making properties of potato, corn and mung bean starches. *J Food Sci Technol* 52:8113–8121. <https://doi.org/10.1007/s13197-015-1954-5>
- Kaur A, Shevkani K, Katyal M et al (2016) Physicochemical and rheological properties of starch and flour from different durum wheat varieties and their relationships with noodle quality. *J Food Sci Technol* 53:1–12. <https://doi.org/10.1007/s13197-016-2202-3>
- Kumar R, Ghoshal G, Goyal M (2019) Synthesis and functional properties of gelatin/CA-starch composite film: excellent food packaging material. *J Food Sci Technol* 56:1954–1965
- Lin JH, Pan CL, Hsu YH et al (2012) Influence of moisture content on the degradation of waxy and normal corn starches acid-treated in methanol. *Food Hydrocoll* 26:370–376. <https://doi.org/10.1016/j.foodhyd.2011.02.020>
- Lutfi Z, Nawab A, Alam F, Hasnain A (2017) Morphological, physicochemical, and pasting properties of modified water chestnut (*Trapabispinosa*) starch. *Int J Food Prop* 20:1016–1028. <https://doi.org/10.1080/10942912.2016.1193514>
- Ma W, Rokayya S, Xu L et al (2018) Physical-chemical properties of edible film made from soybean residue and citric acid. *J Chem* 2018:8
- Mei J, Yuan Y, Wu Y, Li Y (2013) Characterization of edible starch-chitosan film and its application in the storage of Mongolian cheese. *Int J Biol Macromol* 57:17–21. <https://doi.org/10.1016/j.ijbiomac.2013.03.003>
- Mei JQ, Zhou DN, Jin ZY et al (2015) Effects of citric acid esterification on digestibility, structural and physicochemical properties of cassava starch. *Food Chem* 187:378–384. <https://doi.org/10.1016/j.foodchem.2015.04.076>
- Menzel C, Olsson E, Plivelic TS et al (2013) Molecular structure of citric acid cross-linked starch films. *Carbohydr Polym* 96:270–276. <https://doi.org/10.1016/j.carbpol.2013.03.044>
- Oleyaei SA, Almasi H, Ghanbarzadeh B, Moayedi AA (2016) Synergistic reinforcing effect of TiO₂ and montmorillonite on potato starch nanocomposite films: thermal, mechanical and barrier properties. *Carbohydr Polym* 152:253–262. <https://doi.org/10.1016/j.carbpol.2016.07.040>
- Pornsuksomboon K, Holló BB, Szécsényi KM, Kaewtatip K (2016) Properties of baked foams from citric acid modified cassava starch and native cassava starch blends. *Carbohydr Polym* 136:107–112. <https://doi.org/10.1016/j.carbpol.2015.09.019>
- Punia R, Sharma MM, Kalita D et al (2017) Physicochemical, morphological, thermal and pasting characteristics of starches from moth bean (*Vigna aconitifolia*) cultivars grown in India: an underutilized crop. *J Food Sci Technol* 54:4484–4492. <https://doi.org/10.1007/s13197-017-2930-z>
- Saliu OD, Olatunji GA, Olosho AI et al (2018) Barrier property enhancement of starch citrate bioplastic film by an ammonium-thiourea complex modification. *J Saudi Chem Soc* 23:141–149
- Sharma L, Sharma HK, Saini CS (2017) Edible films developed from carboxylic acid cross-linked sesame protein isolate: barrier, mechanical, thermal, crystalline and morphological properties. *J Food Sci Technol* 55:532–539. <https://doi.org/10.1007/s13197-017-2962-4>
- Singh H, Thakur S, Mukhrjee J et al (2017) Influence of acid hydrolysis on physico-chemical, structural, and pasting properties of moth bean (*Vigna aconitifolia*) starch. *Starch/Staerke* 69:1–10. <https://doi.org/10.1002/star.201600242>
- Thessrimuang N, Prachayawarakorn J (2018) Characterization and properties of high amylose mung bean starch biodegradable films cross-linked with malic acid or succinic acid. *J Polym Environ* 27:234–244
- Wang S, Ren J, Li W et al (2014) Properties of polyvinyl alcohol/xylan composite films with citric acid. *Carbohydr Polym* 103:94–99. <https://doi.org/10.1016/j.carbpol.2013.12.030>
- Wani IA, Sogi DS, Hamdani AM et al (2016) Isolation, composition, and physicochemical properties of starch from legumes: a review. *Starch/Staerke* 68:834–845. <https://doi.org/10.1002/star.201600007>
- Xu H, Canisag H, Mu B, Yang Y (2015) Robust and flexible films from 100% starch cross-linked by biobased disaccharide derivative. *ACS Sustain Chem Eng* 3:2631–2639. <https://doi.org/10.1021/acssuschemeng.5b00353>
- Zhou J, Tong J, Su X, Ren L (2016) Hydrophobic starch nanocrystals preparations through crosslinking modification using citric acid. *Int J Biol Macromol* 91:1186–1193
- Zhu F (2015) Isolation, composition, structure, properties, modifications, and uses of yam starch. *Compr Rev Food Sci Food Saf* 14:357–386. <https://doi.org/10.1111/1541-4337.12134>
- Zobel HF, Young SN, Rocca LA (1988) Starch gelatinization: an x-ray diffraction study. *Cereal Chem* 65:443–446

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